



## INTERNATIONAL APPLICATION PUBLISHED UNDER THE PATENT COOPERATION TREATY (PCT)

(51) International Patent Classification <sup>6</sup> : <b>H01M 10/40, 10/04</b>	<b>A3</b>	(11) International Publication Number: <b>WO 99/19917</b> (43) International Publication Date: <b>22 April 1999 (22.04.99)</b>
<p>(21) International Application Number: <b>PCT/EP98/06394</b></p> <p>(22) International Filing Date: <b>8 October 1998 (08.10.98)</b></p> <p>(30) Priority Data: <b>197 44 660.4      9 October 1997 (09.10.97)      DE</b></p> <p>(71) Applicants (for all designated States except US): <b>BASF AKTIENGESELLSCHAFT (DE/DE); D-67056 Ludwigshafen (DE). GS JAPAN STORAGE (JP/JP); Battery Co., Ltd., 1 Inobaba-cho Nishinosho Kisshoin, Minami-ku, Kyoto 601 (JP).</b></p> <p>(72) Inventors; and (75) Inventors/Applicants (for US only): <b>BAUER, Stephan (DE/DE); Hauptstrasse 65a, D-67126 Hochdorf-Assenheim (DE). BRONSTERT, Bernd (DE/DE); Zanderstrasse 35, D-67166 Otterstadt (DE). MÖHWALD, Helmut (DE/DE); Markwardstrasse 16, D-76855 Annweiler (DE). STEPHAN, Oskar (DE/DE); Kolpingstrasse 1, D-68766 Hockenheim (DE). TUKAMOTO, Hisashi (JP/JP); 1, Inobanba-cho, Nishinosho, Kisshoin, Minami-ku (JP).</b></p> <p>(74) Agent: <b>ISENBRUCK, Günter; Bardehle et al, Theodor-Heuss-Anlage 12, D-68165 Mannheim (DE).</b></p>	<p>(81) Designated States: <b>AL, AU, BG, BR, BY, CA, CN, CZ, GE, HU, ID, IL, JP, KR, KZ, LT, LV, MX, NO, NZ, PL, RO, RU, SG, SI, SK, TR, UA, US.</b> Eurasian patent (AM, AZ, BY, KG, KZ, MD, RU, TJ, TM). European patent (AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE).</p> <p>Published <i>With international search report.</i></p> <p>(58) Date of publication of the international search report: <b>24 June 1999 (24.06.99)</b></p>	
(54) Title: <b>COMPOSITE SUITABLE FOR USE IN ELECTROCHEMICAL CELLS</b>		
<p>(57) Abstract</p> <p>A composite comprises Aa) at least one first layer which comprises a mixture Ia, comprising a mix IIa consisting of a) from 1 to 95 % by weight of a solid III, preferably a basic solid III, having a primary particle size of from 5 nm to 20 µm and b) from 5 to 99 % by weight of a polymeric composition IV obtainable by polymerization of b1) from 5 to 100 % by weight, based on the composition IV, of a condensation product V of α) at least one compound VI which is able to react with a carboxylic acid or a sulfonic acid or a derivative or a mixture of two or more thereof, and β) at least 1 mol per mol of the compound VI of a carboxylic acid or sulfonic acid VII which contains at least one free-radically polymerizable functional group, or a derivative thereof or a mixture of two or more thereof, and b2) from 0 to 95 % by weight, based on the composition IV, of a further compound VIII having a mean molecular weight (number average) of at least 5000 and polyether segments in the main chain or a side chain, where the proportion by weight of the mix IIa in the mixture Ia is from 1 to 100 % by weight, and the layer is free of an electron-conducting, electrochemically active compound, and B) at least one second layer which comprises an electron-conducting, electrochemically active compound, wherein the first layer or layers and the second layer or layers are joined to one another by one of the two methods V1 or V2: V1) Lamination of the first layer or layers with the second layer or layers under the action of heat or pressure or under the action of heat and pressure, or V2) Corona treatment of the first layer or layers, the second layer or layers or the first layer or layers and the second layer or layers and subsequent bringing together of the corona-treated first layer or layers with the corona-treated or untreated second layer or layers.</p>		

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Patent- u. Rechtsanwälte  
Galileiplatz 1, München

27. Jan. 2000

Frist  
Bear.

PCT

From the  
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:

ISENBRUCK, Günter  
BARDEHLE et al.  
Theodor-Heuss-Anlage 12  
D-68165 Mannheim  
ALLEMAGNE

Patent- u. Rechtsanwälte  
Mannheim

20. JAN. 2000

Frist: 09.02.2000  
Bear.:

## NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Rule 71.1)

Date of mailing  
(day/month/year)

19. 01. 00

Applicant's or agent's file reference

NAE19970565PC 13/R1

### IMPORTANT NOTIFICATION

International application No.  
PCT/EP98/06394

International filing date (day/month/year)  
08/10/1998

Priority date (day/month/year)  
09/10/1997

Applicant

BASF AKTIENGESELLSCHAFT

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

#### 4. REMINDER

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

Name and mailing address of the IPEA/



European Patent Office  
D-80298 Munich  
Tel. +49 89 2399 - 0 Tx: 523656 epmu d  
Fax: +49 89 2399 - 4465

Authorized officer

Schulz, A

Tel. +49 89 2399-8112



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## PCT

### INTERNATIONAL PRELIMINARY EXAMINATION REPORT



(PCT Article 36 and Rule 70)

Applicant's or agent's file reference <b>NAE19970565P</b>	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. <b>PCT/EP98/06394</b>	International filing date (day/month/year) <b>08/10/1998</b>	Priority date (day/month/year) <b>09/10/1997</b>
International Patent Classification (IPC) or national classification and IPC <b>H01M10/40</b>		
Applicant <b>BASF AKTIENGESELLSCHAFT</b>		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 4 sheets, including this cover sheet.  
  
☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).  
  
 These annexes consist of a total of 6 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☐ Certain observations on the international application

Date of submission of the demand  <b>28/04/1999</b>	Date of completion of this report <b>19.01.00</b>
Name and mailing address of the international preliminary examining authority:   <b>European Patent Office</b> <b>D-80298 Munich</b> <b>Tel. +49 89 2399 - 0 Tx: 523656 epmu d</b> <b>Fax: +49 89 2399 - 4465</b>	Authorized officer  <b>Del Piero, G</b>  Telephone No. <b>+49 89 2399 8579</b>  

# INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/EP98/06394

## I. Basis of the report

1. This report has been drawn on the basis of (*substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.*):

### Description, pages:

1-46 as originally filed

### Claims, No.:

1-12 as received on 17/12/1999 with letter of 17/12/1999

2. The amendments have resulted in the cancellation of:

- ☐ the description, pages:  
☐ the claims, Nos.:  
☐ the drawings, sheets:

3. ☐ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

4. Additional observations, if necessary:

## V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement

### 1. Statement

Novelty (N)	Yes:	Claims	1-12
	No:	Claims	
Inventive step (IS)	Yes:	Claims	1-12
	No:	Claims	
Industrial applicability (IA)	Yes:	Claims	1-12
	No:	Claims	

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EXAMINATION REPORT**

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**2. Citations and explanations**

**see separate sheet**

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT - SEPARATE SHEET**

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International application No. PCT/EP98/06394

The present claims do not introduce matter extending beyond the content of the application as originally filed.

Although formulated in rather broad terms, the subject-matter of the current claims is not anticipated by any of the available citations, which either do not disclose the combination of all the ingredients of the composites according to the present claims or do not mention the combination of at least two layers forming the composites, as defined in the claims, by means of lamination under heat and/or pressure or corona treatment.

The provision of an alternative polymeric, filler-containing electrolyte composite system showing higher mechanical stability than conventional polymeric, filler-containing electrolytes under the stress conditions arising in the production of batteries can form the basis for the acknowledgment of an inventive step.

**Amended set of claims for further prosecution  
(clean copy)**

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We claim:

10 1. A composite comprising

Aa) at least one first layer which comprises a mixture Ia, comprising a mix IIa consisting of

15 a) from 1 to 95 % by weight of a solid III, preferably a basic solid III, having a primary particle size of from 5 nm to 20  $\mu$ m and

20 b) from 5 to 99 % by weight of a polymeric composition IV obtainable by polymerization of

b1) from 5 to 100 % by weight, based on the composition IV, of a condensation product V of

25  $\alpha$ ) at least one compound VI which is able to condense with a carboxylic acid or a sulfonic acid as defined in  $\beta$  or a derivative or a mixture of two or more thereof, and

30  $\beta$ ) at least 1 mol per mol of the compound VI of a carboxylic acid or sulfonic acid VII which contains

AMENDED SHEET

at least one free-radically polymerizable functional group, or a derivative thereof or a mixture of two or ore thereof,

and

5                   b2)   from 0 to 95 % by weight, based on the composition IV, of a further compound VIII having a mean molecular weight (number average) of at least 5000 and polyether segments in the main chain or a side chain,

10                   where the proportion by weight of the mix IIa in the mixture Ia is from 1 to 100 % by weight,

15                   and the layer is free of an electron-conducting, electrochemically active compound,

and

20                   B)   at least one second layer which comprises a polymeric binder and an electron-conducting, electrochemically active compound,

wherein the first layer or layers and the second layer or layers are joined to one another by one of the two methods V1 or V2:

25                   V1)   Lamination of the first layer or layers with the second layer or layers under the action of heat or under the action of heat and pressure, or

30                   V2)   Corona treatment of the first layer or layers, the second layer or layers or the first layer or layers and the second layer or layers and

AMENDED SHEET



subsequent bringing together of the corona-treated first layer or layers with the corona-treated second layer or layers.

2. A composite comprising

Ab) at least one first layer which comprises a mixture Ib comprising a mix IIb consisting of

a) from 1 to 95 % by weight of a solid III, preferably a basic solid, having a primary particle size of from 5 nm to 20  $\mu$ m and

b) from 5 to 99 % by weight of a polymer IX obtainable by polymerization of

b1) from 5 to 75 % by weight, based on the polymer IX, of a free-radically polymerizable compound X selected from the group consisting of

olefinic hydrocarbons, (meth)acrylonitrile, halogens containing olefinic compounds, vinyl alcohol, vinyl acetate, N-vinylpyrrolidone, N-vinylimidazole, vinyl formamide, phosphonitrilic chlorides and derivatives thereof which are partly or completely substituted by alkoxy, phenoxy, amino and fluoroalkoxy groups, aromatic olefinic compounds and vinyl ethers, and which is different from the carboxylic acid or the sulfonic acid VII or a derivative thereof, or a mixture of two or more thereof,

and

AMENDED CLAIMS

b2) from 25 to 95 % by weight, based on the polymer IX, of a further compound VIII having a mean molecular weight (number average) of at least 5000 and polyether segments in the main chain or a side chain,

where the proportion by weight of the mix Ib is from 1 to 100 % by weight

and the layer is free of an electron-conducting, electrochemically active compound,

and

B) at least one second layer which comprises an electron-conducting, electrochemically active compound,

wherein the first layer or layers and the second layer or layers are joined to one another by one of the two methods V1 or V2:

V1) Lamination of the first layer or layers with the second layer or layers under the action of heat or under the action of heat and pressure, or

V2) Corona treatment of the first layer or layers, the second layer or layers or the first layer or layers and the second layer or layers and subsequent bringing together of the corona-treated first layer or layers with the corona-treated or untreated second layer or layers.

3. A composite comprising

at least one first layer Aa or at least one first layer Ab or at least one first layer Aa and at least one first layer Ab,

at least one second layer B,  
each as defined in claim 1 or 2, and

C) at least one bonding layer.

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4. A composite as claimed in claim 3, wherein the bonding layer or layers C has/have a melting point which is lower than the melting point of the first layer or layers or the second layer or layers or the first and second layer or layers.

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5. A composite as claimed in claim 3 or 4, wherein the bonding layer or layers C is/are a polyethylene oxide, a polyvinyl ether, a polyacrylate, a polymethacrylate, polyvinylpyrrolidone, a polyurethane, a wax-like (co)polyolefin, a rubber-like material, polyisobutylene or a mixture of two or more thereof.

15

6. A composite as claimed in any of claims 3 to 5, wherein the bonding layer or layers C comprise(s) a solid III, a plasticizer or a combination of two or more thereof.

20

7. A process for producing a composite as claimed in any of claims 1 to 6, which comprises joining the first layer or layers and the second layer or layers and, if present, the bonding layer or layers to one another by hot lamination.

25

8. A process for producing a composite as claimed in claim 1 or 2, which comprises subjecting the first layer or layers or the second layer or layers or the first layer or layers and the second layer or layers to a corona treatment and subsequently joining the first corona-treated layer or layers to the second corona-treated or untreated layer or layers.

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